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Evaluation of Control Strategies for Volatile Organic Compounds in Indoor Air

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EVALUATION OF CONTROL STRATEGIES FOR VOLATILE ORGANIC COMPOUNDS IN INDOOR AIR

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INTRODUCTION

The Air and Energy Engineering Research Laboratory of the U.S. Environmental Protection Agency (U.S. EPA) conducts and sponsors research on technology to reduce or eliminate emissions of potentially toxic volatile organic compounds (VOCs) from industrial/commercial sources. The realization that indoor air environments, where most humans spend the majority of their time, also contain significant levels of VOCs has led to a number of EPA sponsored studies on monitoring, modeling, and controlling these compounds. The purpose of this paper is twofold: (1) to present an overview of the problem of indoor air pollution by VOCs and approaches to solving it and (2) to report the results of a study to determine the feasibility of carbon adsorption for control of VOCs in indoor air.

BACKGROUND

Sources

There are a wide variety of sources of VOCs in indoor air. In some cases, contributions from outdoor air can be an important component through leakage and makeup air in ventilation systems. Indoor sources vary tremendously depending on the nature of the facility, and are too diverse for any simple classification. However, some broad categories may be identified [1].

- Combustion - Gas fired appliances and kerosene heaters are typically not vented and can contribute to indoor levels of VOCs.
- Smoking - Smoking is probably the most visible source of indoor air pollutants. A number of heavy organics such as naphthalene, aniline, pyridine, and acrolein are emitted along with noxious gases.

- Building Materials - Formaldehyde from insulation applications is an important pollutant in this class. Emission rates of VOCs from materials of construction are of particular importance and have been the subject of a number of recent studies [2].

- Office Machines - Photocopying machines, printers and other machines in use in office environments may be significant sources of VOCs because of the nature of the chemicals needed for their operation.

- Other Sources - Cleaning compounds, adhesives, furnishings, paints, and human activity are all potential sources of toxic pollutants.

There is no database that we are aware of that lists typical indoor concentrations of the different VOCs in diverse environments. However, based on work performed by the Research Triangle Institute for the EPA [3] and some other sources in the literature [4, 5], it appears that these concentrations rarely exceed a few ppm and most often range in the few hundred ppb.

Various studies of indoor air quality (IAQ) have identified more than 250 different organics at levels greater than 1 ppb [6]. Many hundreds of additional compounds undoubtedly exist at lower levels. An evaluation of the data reported indicates the following:

1. An extremely wide variety of organic compounds are found in the indoor environment.

2. The range of measured concentrations between different organic compounds is extremely wide, often two or more orders of magnitude; also, the range of concentrations for a specific compound can vary widely between measurements.

3. In most cases, the concentrations of specific organic compounds exceed the outdoor concentration, thus indicating that the source of the compound is indoors.

4. The sources of the organic compounds are quite numerous within any indoor environment; the sources vary depending on the type of building studied.

While the available IAQ data present a complex picture regarding organic vapors, there is substantial evidence which relates indoor occurrence to emission sources. Table 1 lists some specific indoor sources of organic vapors.

TABLE 1. SPECIFIC INDOOR SOURCES OF ORGANIC VAPORS [2]

Compound	Material Source(s)
Paradichlorobenzene	Moth crystals, room deodorants
Methylene Chloride	Paint removers
Chloroform	Chlorinated water
Formaldehyde	Pressed wood products, foam insulation (UFFI), textiles, disinfectants
Styrene	Plastics, paints
Toluene Diisocyanate	Polyurethane foam, aerosols
Phthalic Acid Anhydride, Trimellitic Acid, Triethylene Tetraamine	Epoxy resins
Sodium Dodecyl Sulfate	Carpet shampoo
Benzyl Chloride, Benzal Chloride	Vinyl tiles plasticized with butyl benzyl phthlate
Ethylene Oxide	Sterilizers (hospitals)

Controls

In evaluating the feasibility of different control strategies, a clear idea of the maximum allowable level for each pollutant is necessary. Unfortunately, toxicity data are lacking for many of the compounds present in indoor air environments. Acceptable air quality for nonoccupational settings is generally based on outdoor ambient air standards or specified indoor ventilation rates. Current control of indoor air quality for organics is often based on reducing levels to those below the odor threshold. Depending on the compound, this may or may not be adequate from a health effects standpoint.

Source removal and ventilation have received the most attention as control strategies. Obviously, source removal is not always practical; for example, offices need photocopy machines to be readily accessible. The development of more sophisticated indoor air flow models will provide information on the effect of source relocation on pollutant levels. This may enable some mitigation at relatively low cost.

Ventilation

Increasing the ventilation rate is another method that has been proposed for indoor air pollution control - another instance of the "dilution is the solution to pollution" theory. Unfortunately, in this case, increased ventilation rates can lead to increased energy usage, making this an economically infeasible option.

Material/Product Selection

The levels of organic vapors in the indoor environment can be affected by the selection of materials and products. Unfortunately, most indoor products have not been sufficiently characterized to allow such selection to occur.

Material/Product Use

The manner in which products are used can impact indoor organic vapor concentrations. Emissions of formaldehyde from pressed wood products decline with time [7], so aging or conditioning of such products prior to installation would reduce emissions. Solvent-containing materials (e.g., paint, adhesives, caulks, paint removers, and waxes) should be used in well ventilated areas. Manufacturers' instructions should be followed. Some activities, such as hobbies, woodworking, and paint stripping, which use high-emission products should be isolated and separate exhaust fans provided. Solvents and solvent-containing products should be stored in airtight containers; outside storage is preferred.

Other Measures

Other measures for controlling the levels of indoor organic vapors include:

- Building "bake out," the process by which new or renovated buildings are heated and ventilated prior to occupancy [8]; and
- Formaldehyde removal via fumigation.

It is apparent from the above discussion that a need exists for developing practical methods for the removal of organic vapors from indoor air environments. Adsorption, absorption, incineration, and catalytic conversion are all removal techniques for pollutant gases. All have demonstrated applicability in conventional pollution control applications. A recent study by Collins [9] reports on a low-temperature catalyst used in a room air filtration device. However, adsorption on activated carbon seems to be the most popular technique currently in use for indoor applications. A review by Turk [10] discusses in detail the applicability of carbon adsorption to pollution control.

PURPOSE OF THE STUDY

A recent study by the Research Triangle Institute for the EPA concluded that the majority of adsorption systems currently being marketed for indoor air pollution applications dealt with odor control. Quantifiable performance data were not available for any of the systems surveyed. The carbon in each unit was assumed to be spent when occupant complaints increased. Obviously, this method of bed lifetime estimation has drawbacks, particularly when dealing with potentially toxic compounds that have relatively high odor thresholds. The study also found that adsorption isotherm data in the low concentration regime (<10 ppm) were not publicly

available for even simple compounds on any carbons. This led to our effort to obtain adsorption isotherms in the ppb range in order to determine the applicability of activated carbon adsorption for indoor air pollution control.

EXPERIMENTAL DESIGN

Three types of activated carbon were tested: (1) GAC 410G, a coal-based carbon manufactured by the Ceca Division of Atochem, Inc., (2) AC-35, a wood-based carbon from the same manufacturer, and (3) a type AC coconut-shell-based carbon manufactured by Barneby-Cheney. Table 2 lists some of the physical characteristics of these carbons.

The pollutants chosen were relatively simple, low molecular weight compounds representative of three different classes of organic compounds: benzene (aromatic hydrocarbon), acetaldehyde (oxygenated hydrocarbon), and 1,1,1-trichloroethane (halogenated hydrocarbon). Isotherms were measured in the 100 to 200 ppb range for each compound.

Materials and Equipment

The low concentrations were generated with clean, dry air streams flowing over permeation tubes with certified permeation rates. These tubes were obtained from Vici Metronics and had permeation rates of 71.3 ng/min for benzene, 148 ng/min for 1,1,1-trichloroethane, and 32.5 ng/min for acetaldehyde. Nitrogen was used as a purge gas during degassing. All gases used had purities greater than 99.999%.

The experimental setup is shown in Figure 1. It consisted of two mass flow controllers with flow ranges of 0-1000 cm^3/min and 0-100 cm^3/min, a Cahn Instruments Model 1000 microbalance, and a thermostatted water bath for the permeation tubes and balance jacket.

TABLE 2. CARBON PROPERTIES

Property \ Carbon	CECA GAC 410G	CECA AC-35	Barneby-Cheney AC
Base Material	Coal	Pinewood Charcoal	Coconut Shell
Activation Method	Steam	Steam	Steam
CCl_4 Adsorption Wt. % (min)	60	65	60
Surface Area m^2/g	1050-1150	1150	1050-1150

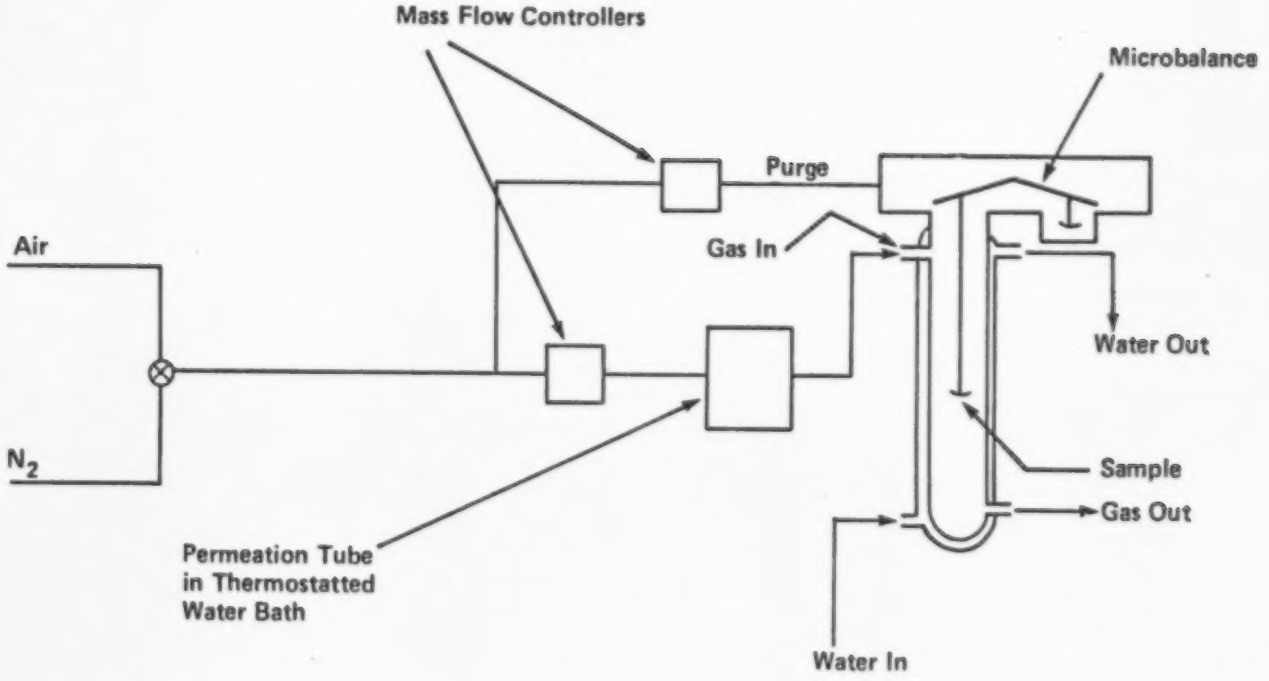

Figure 1. Schematic of experimental setup.

The Cahn model 1000 microbalance was used for the adsorption studies. It is capable of weighing up to 100 g with an accuracy of ± 0.5 ug. This device may be operated under low temperature, high vacuum conditions for the determination of BET surface area and is ideally suited to make adsorption isotherm measurements under different conditions of pressure and temperature.

Experimental Procedure

Gravimetric measurement of adsorption isotherms can be carried out in either a static or dynamic mode. The static mode, usually under a vacuum, has the advantages of a high signal-to-noise ratio and a fairly good control of partial pressure of adsorbate. However, equilibration times can be very long, particularly at low concentrations, and the conditions are not representative of real systems, where air is present with the adsorbate and may affect its adsorptivity. We chose to operate in the dynamic (flow) mode, at atmospheric pressure, in order to reduce experimental time and measure adsorption under conditions more representative of streams challenging real world filters.

The carbon sample (150 to 200 mg) was degassed under flowing nitrogen while it was heated with an infrared lamp. Degassing was assumed to be complete when the weight remained constant for 10 to 15 minutes. The sample was then allowed to cool in nitrogen before switching to the air stream and introducing the permeation tube. Flow rates were adjusted to give the desired concentrations. Equilibrium was assumed when the weight gain (on the 100 ug scale) levelled off.

RESULTS

Table 3 summarizes the results for the uptakes of organic pollutants on the various carbons. It is apparent that the capacities (mol/g carbon) do not vary much with either compound or carbon in the concentration range studied. Capacities ranged from 10^{-8} to 10^{-6} mol/g. Acetaldehyde uptake is about 2-3

times lower than the other compounds studied. Figures 2, 3, and 4 represent the isotherms over the concentration ranges studied. The magnitude of isotherm slopes in the concentration range studied decreased from benzene to 1,1,1-trichloroethane to acetaldehyde. Higher isotherm slopes indicate more favorable adsorption. The data for benzene, for which three data points are available, were linearized in order to obtain an equation for use in a predictive packed bed model. Although the data (Figure 2) do not appear to fit a straight line very well, we felt that this exercise would give us a feel for the applicability of carbon adsorption to indoor air pollution control. Table 4 lists the linearized isotherms for benzene adsorption on the three carbons in this study.

Isotherm data are useful for carbon screening but do not provide direct practical information on the applicability of the technique. For this reason, an estimate of carbon filter size for benzene (which probably represents the best case for the compounds studied) was carried out using a model developed at RTI [11]. The linearized form of the equation on Barneby-Cheney carbon was used. The challenge concentration of benzene was assumed to be 150 ppb, and the superficial velocity of gas across the filter was assumed to be 12.7 cm/sec, corresponding to a flow of 100 cfm (283 lpm) across a 2 x 2 ft (61 x 61 cm) filter. An exit concentration of 50 ppb was set as the level corresponding to useful bed life. Table 5 lists the results of this simulation. It is apparent that breakthrough occurs rather quickly, bringing into question the applicability of a once-through filter. A 6-in. (15 cm) thick filter, such as that typically marketed for odor control applications, would break through almost immediately after installation, even in the absence of other pollutants or humidity effects, both of which would cause performance to deteriorate further. It is also important to note that the level at which acceptable breakthrough is set is critical to bed lifetime. In the absence of

TABLE 3. SUMMARY OF ADSORPTION RESULTS

Compound		Uptake*	
Acetaldehyde	AC-35	GAC 410G	Barneby-Cheney
Concentration (ppb)	mol CH_3CHO/g C	mol CH_3CHO/g C	mol CH_3CHO/g C
119	3.39E-07	3.52E-07	1.99E-06
153	4.17E-07	1.00E-06	1.00E-06
Benzene	AC-35	GAC 410G	Barneby-Cheney
Concentration (ppb)	mol C_6H_6/g C	mol C_6H_6/g C	mol C_6H_6/g C
101	2.42E-07	1.24E-07	4.30E-07
119	4.30E-07	6.37E-07	5.60E-07
176	1.00E-07	7.27E-07	6.33E-07
1,1,1-Trichloroethane	AC-35	GAC 410G	Barneby-Cheney
Concentration (ppb)	mol CH_3CCl_3/g C	mol CH_3CCl_3/g C	mol CH_3CCl_3/g C
115	7.49E-08	1.15E-07	2.04E-07
183	1.33E-07	2.80E-07	1.91E-07

* each number represents the average of 3-4 data points

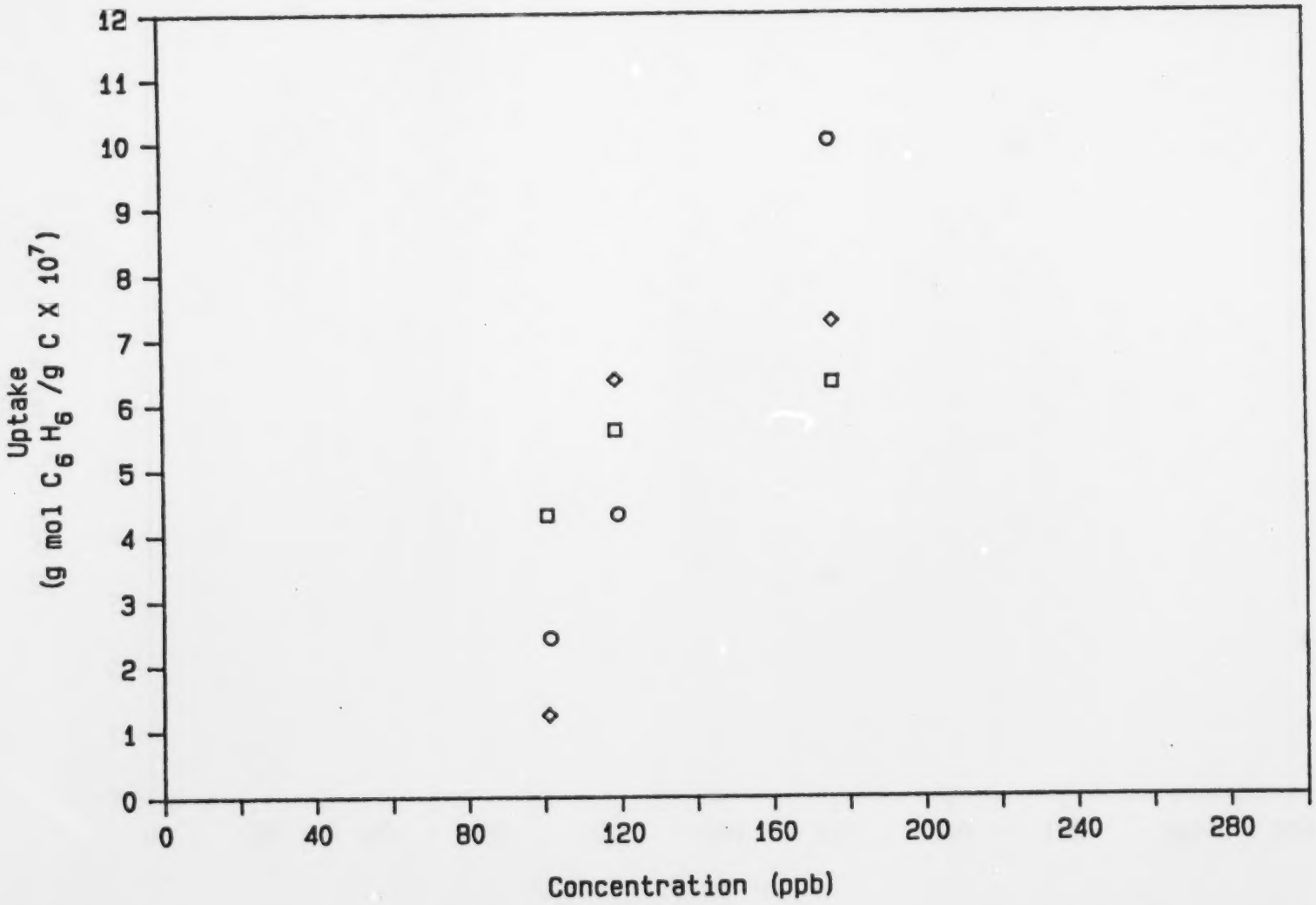

Figure 2. Adsorption of benzene on activated carbons.

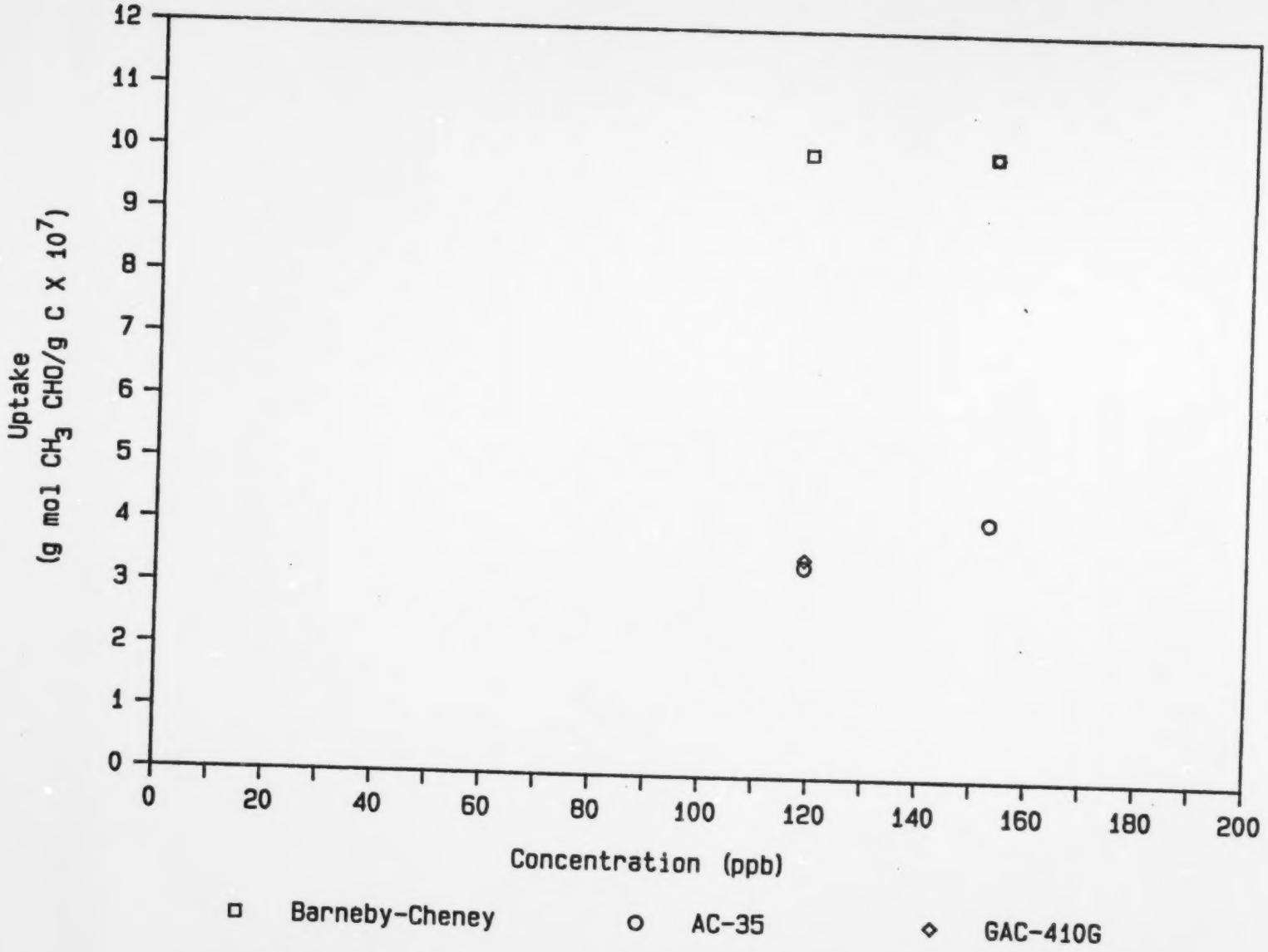

Figure 3. Adsorption of acetaldehyde on activated carbons.

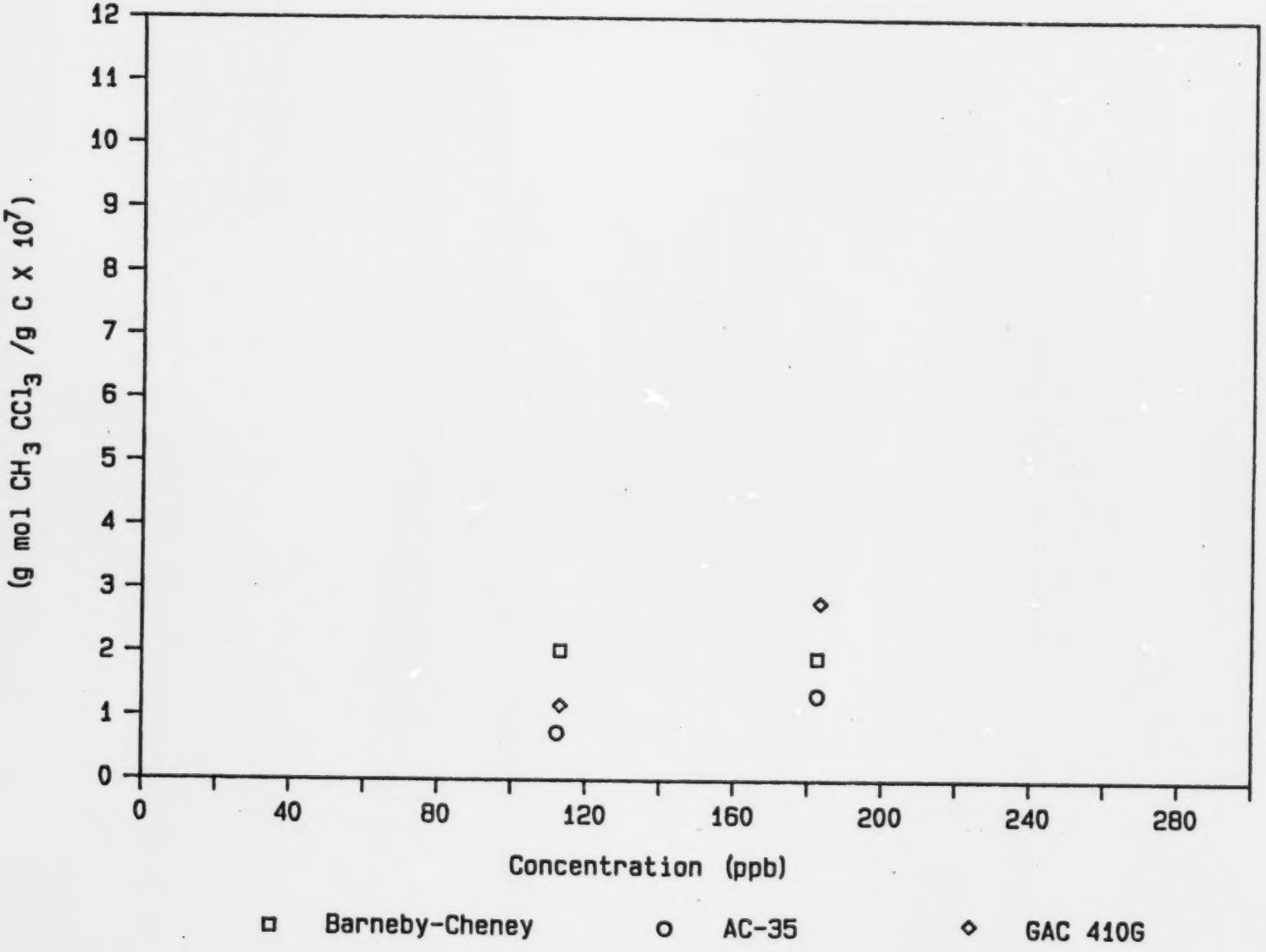

Figure 4. Adsorption of 1, 1, 1-trichloroethane on activated carbons.

toxicological data, this may have to be set rather arbitrarily, erring on the side of caution. More complex flow schemes, involving parallel beds with regeneration, may work, but may be uneconomical except in very large scale applications. Carbon adsorption may be viable in the event of major surges in concentration such as spills or releases, which push the ambient concentrations into regimes where the shape of the isotherm is more favorable and sufficient driving force exists for a reasonable uptake.

CONCLUSIONS

The following conclusions may be drawn from this study:

1) All of the carbons studied showed measurable uptakes of benzene, acetaldehyde, and 1,1,1-trichloroethane.

2) The adsorption of all the compounds on the three carbons was similar, in the range of 10^{-8} to 10^{-6} gmol/g.

3) A packed bed simulation showed that breakthrough occurs very quickly in the shallow (6-in. or 15 cm) filters currently marketed for odor control.

4) Carbon adsorption may be applicable for "event control"; i.e., sudden surges of concentration caused by spills or leaks.

5) Alternative techniques such as low-temperature catalytic oxidation or noncatalytic oxidation over $KMnO_4$ impregnated pellets need to be investigated for continuous control applications.

TABLE 4. LINEAR EQUATIONS FOR BENZENE.*

Carbon Type	
AC-35:	uptake = 1.0×10^{-8} C_b -7.7×10^{-7} mol C_6H_6/g C
GAC 410G	uptake = 6.5×10^{-9} C_b -3.6×10^{-7} mol C_6H_6/g C
Barneby-Cheney	uptake = 2.4×10^{-9} C_b $+2.3 \times 10^{-7}$ mol C_6H_6/g C

* C_b = benzene concentration in ppb

TABLE 5. VARIATION OF EXIT CONCENTRATION WITH BED DEPTH

T = 4320 hr (6 mos)		T = 30 hr	
L (ft)*	C exit (ppb)	L (ft)*	C exit (ppb)
1098.64	121	7.65	114
1097.67	115	8.15	55.6
1108.95	99.5	8.31	35.1
1137.29	67.2	8.41	20
1163.09	40.6		

Compound: Benzene

150 ppb challenge concentration
12.7 cm/s superficial velocity
3.0 mm particle size
Barneby-Cheney Type AC Carbon

* 1 ft = 0.3048 m

REFERENCES

1. Wadden, R.A. and Scheff, P.A. 1983. Indoor Air Pollution: Characterization, Prediction and Control, John Wiley & Sons.

2. U.S. EPA, June 1987. EPA Indoor Air Quality Implementation Plan: Appendix A. Preliminary Indoor Air Pollution Information Assessment, EPA/600/8-87/014.

3. Wallace, L.A., Pellizzari, E., Leaderer, B., Zelon, H., and Sheldon, L., 1987. "Assessing Emissions of Volatile Organic Compounds by Building Materials and Consumer Products," Atmospheric Environment, 21, 385-393.

4. Wang, T.C., 1975. "A Study of Bioeffluents in a College Classroom", ASHRAE Trans., 81 (Part 1):32-44.

5. Tucker, W. Gene, 1986. U.S. EPA/AEERL, Personal Communication.

6. Sterling, D.A., 1985. "Volatile Organic Compounds in Indoor Air: An Overview of Sources, Concentrations, and Health Effects". In: Gammage, R.B., Kaye, S.B., Jacobs, V.A., eds. Indoor Air and Human Health, Chelsea, MI, Lewis Publishers, Inc., pp. 387-402.

7. Hawthorne, A.R., Gammage, R.B., Dudney, C.S., 1984. "Effect of Local Geology on Indoor Radon Levels: A Case Study", 3rd International Conference on Indoor Air Quality and Climate, v. 2, Radon, Passive Smoking, Particulates and Housing Epidemiology, Stockholm, Sweden, Swedish Council for Building Research; pp. 137-148.

8. State of California, 1984. Building Closeout Procedure. Sacramento, CA: Office of the State Architect.

9. Collins, M.F., June 1986. "Characterization of the LTC Catalyst: Performance Against Common Air Pollutants", presented at the 79th Annual Meeting of the Air Pollution Control Association, Minneapolis, MN. Air Pollution Control Association; paper no. 86-43-3.

10. Turk, A., 1977. "Adsorption In: Air Pollution", Stern, A.C., Ed., Vol. IV, 3rd ed., Academic Press, New York.

11. U.S. EPA, February 1987, EAB Control Cost Manual (3rd ed.), EPA-450/5-87-001A.